

## Preface to Special Topic: Invited Articles on Microfluidic Rheology

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This special issue on *Microfluidic Rheology* includes review and original articles focusing on a promising area of research and development, namely, the use of microfluidic technology for rheological measurements or “rheology-on-a-chip” (ROC). It discusses new advances in using ROC technology to control the flow, and hence measure the material properties of a variety of complex fluids, including biological fluids and “active” suspensions. The reader will note that this special issue includes contributions from researchers of various disciplines. This should not come as a surprise — rheology is inherently an interdisciplinary field that brings together academic and industrial researchers spanning the fields of engineering, material science, chemistry, mathematics, physics, geology, and biomedical sciences. From a technological point of view, rheological measurements are routinely performed, often guiding product development and quality control, in many industries including consumer goods (e.g., detergents, cosmetics, and paints), foodstuffs (e.g., chocolate and yogurt), petroleum, chemical, polymer and plastic, and pharmaceuticals. Instrument development has been a key part of the field as understanding flow and rheological properties (viscosity, elastic and shear moduli, etc.) of a material/fluid relies on accurate and precise measurements. Indeed, many excellent rheological instruments with a range of capabilities are commercially available. So why would anyone want to perform rheological experiments using microfluidic technology?

As many of the contributing authors of this special issue argue, there are advantages in using microfluidic technology to measure the rheological properties of fluids. Perhaps the most recognizable benefit of ROC technology is that inertial effects are minimized in microfluidic flows due to the small system length scale  $L$ , usually  $\sim O(10^2)$   $\mu\text{m}$ . This feature can be best understood by considering the Reynolds number ( $Re$ ), which quantifies the relative importance of fluid inertia to viscous forces and is usually defined as  $Re = \rho VL/\mu$ . Here,  $L$  is a characteristic measure of the system size,  $V$  is the characteristic fluid velocity, and  $\rho$  and  $\mu$  are the fluid density and viscosity, respectively. Due to the small system size  $L$ ,  $Re$  can be small ( $<1$ ) even for low viscosity liquids. In fact, one can obtain large velocity gradients or shear-rates ( $V/L \sim 10^4\text{--}10^6$   $\text{s}^{-1}$ ) while still keeping the flow laminar. Another attractive feature of ROC is the ability to directly visualize the fluid microstructure while simultaneously performing a rheological measurement. Such combined measurements are particularly useful in the study of complex fluids (e.g., colloidal suspensions, foams, emulsions, etc.), which possess structure at an intermediate or mesoscale; the non-Newtonian properties that are often observed in complex fluids stem from the flow induced changes of the mesoscale structure. The combination of ROC technology and microscopy offers the possibility to investigate and understand the relationship between the fluid microstructure and its (bulk) macroscopic flow properties. Furthermore, the effects of confinement on both the microstructure and bulk flow behavior can be systematically studied using microfluidic devices. There are, of course, other advantages in using ROC technology including low sample volume, continuous operation, the potential for portability, and price. But there are challenges as well. For example, it is still not a simple task to develop flows with controllable kinematics in microfluidic devices (see Refs. 9, 11, 7, 1, and 5), and while progress has been made, measuring instruments such as pressure and flow rate sensors

are still not sensitive/accurate or small enough to be easily incorporated in ROC platforms. To circumvent these resolution issues, researchers have begun to develop new measurement methods and techniques. For example, the onset of elastic instabilities or particle migration in a complex fluid<sup>4</sup> can be used to estimate the fluid relaxation time while birefringence can be used to access local stress differences making pressure-drop measurements unnecessary.<sup>1</sup> In general, as the reader will see in the special issue, much progress has been made lately and ROC technology is leaving the realm of promise and is becoming a reality.

This issue contains three review articles and nine regular articles, covering a range of key topics and developments including novel ROC designs and exciting new ideas to measure the material properties and understand the flow of complex fluids. The *review articles* cover shear and elongational rheometry as well as the flow and dynamics of DNA molecules in microfluidic devices. Haward<sup>1</sup> provides a comprehensive review of elongational rheometry and flow of complex fluids in microfluidic devices. The article briefly reviews the main methods to obtain the extensional viscosity of fluids and then discusses recent advances in using microfluidic technology for elongational rheometry. The main focus is on the cross-slot geometry, which is characterized by an extensional flow with a hyperbolic point. In particular, the author discusses the use of experimental data and numerical simulations to optimize the geometry of the cross-slot to achieve true planar (2D) extensional flow, as well as the development of oscillatory flows for unsteady measurements. Finally, the development of flow instabilities, which is one of the main limitations of this geometry, is discussed. Shear rheology in microfluidic devices is reviewed by Gupta and co-workers<sup>2</sup> who focus on measurements and flow of complex and biological fluids. The authors discuss different methods of performing shear rheology in microfluidic devices, including perhaps the two most popular techniques, namely, pressure drop and flow sensing. In pressure sensing, a (mass) flow rate is imposed and the goal is to measure the resultant pressure drop across the device, while in flow sensing, a pressure drop is imposed and the resultant flow rate is measured. The pressure sensing method can be thought as analogous to a rate-controlled mode while the volume sensing method can be thought as analogous to a stress-controlled mode. The authors go on to discuss shear rheometry in microfluidic devices using surface-tension, co-flowing streams, diffusion methods as well as techniques based on measuring the fluid velocity profiles. As briefly mentioned above, ROC technology offers the opportunity to directly visualize the fluid microstructure. In our final review, Rems and co-workers<sup>3</sup> discuss some of the main concepts governing the flow of DNA molecules in microfluidic devices as well as applications. DNA molecules have long served as model polymers due to their ease of visualization (fluorescent labeling) and manipulation (size, flexibility, etc.) and have been widely used to study fundamental aspects of polymer physics and fluid dynamics. In particular, the microstructure of DNA solutions can be directly visualized and tracked using fluorescent labeling of DNA molecules, which is one of the advantages of microfluidic technology. The authors review some of the main advances made in polymer physics using DNA molecules to test long-standing predictions as well as the dynamics of DNA molecules in shear, extensional, and other types of flows. The discussion proceeds to the dynamics of DNA molecules under confinement particularly in nano-fluidic systems and through nano-pores. The paper concludes with relevant applications in the manipulation of single molecules that can be used, for example, to inject DNA into cells.

The series of regular articles that follow represent the state-of-the-art in the field. The series begins with a discussion of an often overlooked design parameter in developing ROC technology, namely, the choice of building material. Traditionally, polydimethylsiloxane or PDMS, an elastomer, has been the material of choice for microfluidic fabrication due to its versatility, bio-compatibility, and low cost. But as Del Giudice and co-workers<sup>4</sup> demonstrate, the deformation of PDMS during flow can lead to unusual particle dispersion when compared to channels made with rigid walls. In particular, the authors show that particle migration can be used to obtain the relaxation time of viscoelastic fluids but find that wall stiffness can affect the measurements. This effect, however, can be mitigated by keeping the ratio of the pressure drop  $\Delta P$  to the wall Young's modulus  $E$  small. Following the vein of new techniques, Josephson *et al.*<sup>6</sup> combine particle tracking micro-rheology and microfluidics to measure the viscosity of

protein (antibody) solutions as a function of temperature. They discuss another advantage of microfluidic technology, namely, enhanced heat transport and rapid temperature equilibration. They use a Peltier module for temperature control and are able to systematically investigate the effects of temperature, ranging from 1 °C to 40 °C, on the shear viscosity of protein solutions as a function of antibody concentration. With their set-up up to 72 temperature-concentration measurements can be made in 1 day. It is worth noting that the viscosity of some of these solutions at 40 °C can achieve values of less than 1 mPa s, which is quite difficult to measure in conventional (macroscopic) rheometers. Protein solutions with high viscosity, on the other hand, are investigated by Dhamaraj and co-workers,<sup>12</sup> who measure the viscosity of enzymatic (lysozyme) solutions as a function of shear-rate and temperature using a pressure-driven microcapillary rheometer and scattering. They find that, as expected, the solution viscosity increases with protein concentration and decreases with temperature. However, while at low protein concentration, the viscosity dependence seems to follow an Arrhenius relationship (see also Ref. 6), deviations are found at larger concentration possibly due to the onset of glassy behavior. They also found a curious shear-thinning behavior at high shear-rates.

Keshavarz and McKinley<sup>5</sup> describe two novel devices for measuring the extensional viscosity  $\eta_e$  of fluids, an elusive quantity to measure (see also Ref. 1): (i) an extensional-viscosity-rheometer-on-a-chip or EVROC and (ii) Rayleigh Ohnesorge jetting extensional rheometry or ROJER. While EVROC is a contraction/expansion flow in which *in-situ* pressure-drop measurements are related to extensional viscosity, ROJER relies on capillary breakup of viscoelastic liquid jets. The authors discuss in detail the main physical mechanisms governing EVROC and ROJER and show that they can indeed be used to measure  $\eta_e$  of a range of dilute polymeric fluids; comparisons to well-known models are made to put the measurements in perspective. The authors provide the operating range and limits for both devices as well as a very useful nomogram for extensional rheometry of viscoelastic fluids in general.

Designing and characterizing the flow, single and multiphase, of fluids in microfluidic devices is perhaps one of the most important steps in developing ROC devices. In this special issue, the need to design geometries with a range of flow types is addressed by Dockx and co-workers.<sup>9</sup> The authors use a counter-flowing microfluidic device with a stagnation point in order to vary the amount of vorticity in the flow. Both numerical simulations and confocal microscopy are used to demonstrate that indeed a wide range of flow types can be achieved with only two inlets by manipulating the channel geometry, in particular, the dimensionless gap length and the channel depth to height ratio. The next step would be to perform rheological measurements in the proposed device. Further insights into the flow of viscoelastic fluids in the EVROC device, briefly mentioned above and discussed in detail in Ref. 5, are presented by Zografos and co-workers<sup>11</sup> using numerical simulations for flow optimization. The authors propose simple modifications, guided by simulations, to the EVROC geometry in order to generate a homogenous flow with a constant extensional strain-rate along the centerline for different types of fluids. Both pressure- and electro-osmotic driven flows are investigated. The authors discuss the findings as a function of several geometric (aspect and contraction ratios) and flow parameters ( $Re$  and  $Wi$ ). Next, the effects of shear-thinning viscosity and elasticity on the flow behavior of complex fluids in (model) porous media in a microfluidic device are discussed by Machado and co-workers.<sup>10</sup> These types of flows inform the design of ROC devices and are relevant to many applications in the chemical and petroleum industry (e.g., oil recovery, hydraulic fracturing, etc.). The authors use both numerical simulations and experiments to show a significant enhancement in flow resistance and pressure drop as elastic effects increase ( $Wi \geq 10$ ) and flow instabilities develop; an increase in (mean) flow uniformity, compared to Newtonian fluids, is also observed. Finally, a study on the capillary breakup of liquid jets for the production of water-in-oil drops in a flow-focusing device using AC fields is provided by Castro-Hernandez and co-workers.<sup>7</sup> In their experimental setup, they find that the length of fluid jets can be manipulated with high precision by controlling the applied voltage, frequency, and liquid conductivity. In particular, the authors find a critical voltage threshold in which the jet length increases very rapidly.

Our final contribution is from Alonso-Matilla and co-workers<sup>8</sup> who focus on the flow of “active” particle suspensions in micro-channels. Active suspensions are fluids in which “active” or living particles are present in the fluid medium; they have been the subject of much attention recently. These self-propelling particles can inject energy into the system and generate mechanical stresses and create flows within the fluid medium. This internally injected energy drives the fluid out of equilibrium (even in the absence of external forcing) and can lead to novel properties not seen in passive suspensions. The authors develop a nonlinear mean-field kinetic theory to analyze the flow and dynamics of swimmers, pullers and pushers, in planar Poiseuille flow in micro-channels of varying widths. The role of swimming kinematics (and density) on particle distribution, migration, orientation, and flow resistance is discussed. They find that activity effects dominate in weak flows where pushers and pullers lead to a decrease or increase in fluid viscosity, respectively; these effects vanish for strong flows. These predictions can in the future be directly compared to microfluidic measurements and contribute in this way to a rapidly growing field, in which rheologists can play a significant role.

We are certain the reader will find the reviews and regular articles in this issue interesting and thought-provoking. The present collection of papers discusses the design of novel flow geometries, new understanding of the dynamics of fluid microstructure under flow, and the development of practical rheology-on-a-chip (ROC) devices. We hope that this special issue will stimulate further progress not only in the development of ROC devices but also on the understanding of flow of complex fluids.

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<sup>3</sup>L. Rems, D. Kawale, L. J. Lee, and P. E. Boukany, *Biomicrofluidics* **10**, 043403 (2016).

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<sup>5</sup>B. Keshavarz and G. H. McKinley, *Biomicrofluidics* **10**, 043502 (2016).

<sup>6</sup>L. L. Josephson, W. J. Galush, and E. M. Furst, *Biomicrofluidics* **10**, 043503 (2016).

<sup>7</sup>E. Castro-Hernández, P. García-Sánchez, J. Alzaga-Gimeno, S. H. Tan, J.-C. Baret, and A. Ramos, *Biomicrofluidics* **10**, 043504 (2016).

<sup>8</sup>R. Alonso-Matilla, B. Ezhilan, and D. Saintillan, *Biomicrofluidics* **10**, 043505 (2016).

<sup>9</sup>G. Dockx, T. Verwijlen, W. Sempels, M. Nagel, P. Moldenaers, J. Hofkens, and J. Vermant, *Biomicrofluidics* **10**, 043506 (2016).

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<sup>11</sup>K. Zografos, F. Pimenta, M. A. Alves, and M. S. N. Oliveira, *Biomicrofluidics* **10**, 043508 (2016).

<sup>12</sup>V. L. Dharmaraj, P. D. Godfrin, Y. Liu, and S. D. Hudson, *Biomicrofluidics* **10**, 043509 (2016).